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The heavy metal content of skeletons from an ancient metalliferous polluted area in southern Jordan with particular reference to bioaccumulation and human health

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Abstract

This paper considers pollution/toxicological science in an archaeological context. Copper mining was an important activity in southern Jordan, especially during the Bronze Age, Nabatean, Roman, and Byzantine periods, and the environmental legacy of such intensive mining and smelting activities exists today in the form of massive, ancient spoil and smelting tips. The environment was heavily polluted by copper, lead, and other cations during these early periods and the effects of such pollutants continue into modern times. Samples of goat, sheep, and Bronze Age and Byzantine skeletons have been analyzed and high metal loads, from uptake by diverse processes, are reported. Emphasis is placed on the importance of sampling procedure and sample location, bioaccumulation, and the partitioning of such elements. Implications of such pollutants in terms of environmental and human health in ancient and modern times are discussed. Teeth are found to provide excellent vehicles for the monitoring of pollution in both ancient and recent times. Bronze Age skeletons exhibited chemical fingerprints different from those of the Byzantine period.

Keywords: Jordan; Copper; Lead; Skeleton; Human health; Bioaccumulation; Toxicology

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1. Introduction

In the southern part of the Hashemite kingdom of Jordan, to the north-northeast of the important Red Sea town of Aqaba and some 70 km to the southeast of the Dead Sea, is an isolated stony desert area in which a number of major wadis are located; these include Wadi Feynan, Wadi Ghuwayr, Wadi Shegar, and Wadi Dana. These wadis, during research visits in 2000, 2001, and 2002, were dry. Infrequently they carry water, which has drained from elevated basaltic, limestone, and sandstone areas located primarily to the east and north of the research sites. Wadi Faynan is the location of the notorious Roman copper mining and smelting center based around the ruined city or ancient industrial center of Phaino; here the worst criminals in the Roman Empire were condemned to slavery in the mines and the associated extractive activities. The mines and adits in the Wadi Faynan area run through complex ore bodies, which were produced during several periods of mineralization. Copper, lead, and chromium are well represented, together with other potentially toxic cations such as cadmium. These, as noted by Alloway and Ayres (1993) would be liberated during both quarrying and ore processing. The area is currently inhabited by Bedouin who have scattered encampments; the Bedouin are highly dependent upon their goats and sheep, which they pasture on the limited (low biomass per unit area) vegetation growing in this arid area. The paucity of vegetation is further aggravated by the heavy metal pollution prevalent in this area (Pyatt et al., 2000).

Dramatic features of the landscape of this region are the massive mining spoil and smelting waste tips, which have gradually been undergoing environmental weath-ering/amelioration for a period in excess of two millennia. Indeed, as noted by Pyatt et al. (1999), ''From the Bronze Age and thence through the Iron Age, Nabatean, Roman and Byzantine periods, extensive mining, particularly for copper (and lead), occurred and the legacy of such extensive ancient metallurgical enterprises remains today in the form of major spoil tips where cations including copper, lead and manganese are present in high concentrations in both the spoil tips and the associated sediments.'' Hauptmann et al. (1992) reported that ore exploitation has occurred at Feinan since the seventh millennium BC; they recorded peak values of copper production in the early Bronze Age, the Iron Age, and the Roman period. Additionally, they record that in close proximity to the ruins of the Roman city of Phaino are over 250 copper mines and associated spoil tips, together with extensive slag heaps. The conditions for the slaves would have been appalling, with severe environmental pollution and periodic food shortages, together with strenuous manual labour; indeed, Eusebius of Caesarea (1989) describes how severe were the conditions to which the prisoners were exposed. Eventually mining was to decrease in this area; thus Lewis (1996) noted that industrial-scale copper exploitation appears to have ceased with the expulsion of the Byzantine administration from this area in the seventh century AD (i.e., 1300 years BP).

It is hence apparent that these massive scattered spoil tips have had a prolonged period to undergo processes of environmental weathering, but not necessarily clearly defined amelioration, by diverse processes of atmospheric erosion, leaching, sheet and gulley erosion, etc. (Pyatt and Birch, 1994; Gee et al., 1997). Indeed, Pyatt et al. (2000) note that in both arid and semiarid environments, pollutants produced during ancient times are not generally dissipated to any marked extent. Furthermore, organisms in this area will have been subjected to a toxic environment for a prolonged period; consequently opportunities for adaptation and the development of ecotypes are conceivable.

Stretching away from some of the spoil and smelting tips are the remains of a once extensive field system; this, with the associated irrigation channels, is believed to have been formerly used for food production to sustain the mining and associated military populations inhabiting and exploiting the area in the Nabatean, Roman, and Byzantine periods. However, it does also incorporate traces of even earlier agricultural systems (Barker et al., 1997, 1998, 1999).

This current paper examines some of the effects and implications of the exposure of mammals, including humans, to such ancient and persistent environmental toxicology; additional information, particularly concerning the archaeology and aspects of the environmental sciences of this area, is reported in a number of excellent and comprehensive papers by Barker et al. (1997, 1998, 1999). The current study is also of importance and relevance to modern polluted areas in terms of environmental and human health aspects, as many of the findings resultant from this research can be extrapolated.

Humans and their animals will have been, and continue to be, important recipients of cations (Pyatt et al., 1999, 2000, 2002a; Pyatt and Grattan, 2001); certainly earlier inhabitants acting as slaves in the copper extraction activities would have been exposed to severe pollution episodes (Pyatt et al., 2000), and this arguably would have had adverse effects upon their health. Individuals would have been recipients of copper and lead from the following sources: inhalation and ingestion of airborne particulates, ingestion as a consequence of trophic level accumulation of cations (from food), and the drinking of contaminated water. It should be noted that similar mechanisms would also operate in modern times (Chao and Wang, 1994; Cohen, 1979). However, as indicated by Pyatt et al. (2000, 2002a), differences would occur insofar as overworked slaves in ancient times conceivably would have been less resilient than modern inhabitants of the area and thus prone to a diversity of health problems such as bone damage and exhaustion. Early pollution problems may also have been intensified by malnutrition or starvation as a consequence of poor local crop production on the polluted soils, together with poor medical and diagnostic support. These effects would have been intensified as a result of synergistic/addition effects generated by other pollutants (e.g., sulfur dioxide, carbon monoxide, oxides of nitrogen, and particulates) emanating from activities such as the smelting of copper ore.

2. Some effects of copper and lead on humans

While copper is essential to humans, it can prove toxic in enhanced concentrations. Effects include diarrhea, nausea, vomiting, coma, and ultimately death. Scheinberg (1979) reported that inhalation of dusts of copper salts can lead to perforation of the nasal septum and noted that pulmonary copper deposition can result in the development of lung and liver tumors, while Theophanides and Anastassopoulou (2002) reported that copper is involved in carcinogenicity as catalytic copper.

In urban areas, approximately 30 mg of lead is absorbed daily by humans via the gastrointestinal tract and approximately 20 mg via the respiratory tract (Engel et al., 1971); deposition of lead occurs largely in the skeleton. The effects of lead pollution may be summarized as follows (Pyatt et al., 1999, 2000, 2002a; Pyatt and Grattan, 2001); constipation, decrease in appetite, abdominal cramps, headache, motor nerve paralysis, anaemia, fatigue, and brain damage. Lead has been implicated by Boeckx (1986) in inhibiting heme synthesis and in decreasing red cell survival, in carcinogenicity (Gaffey and Cooper, 1980) and in nucleic acid destabilization by Farkas (1975). Of particular interest in the context of this present research programme is a paper by Gorell et al. (1999) where lead, in combination with other cations such as copper, has been implicated in Parkinson's disease; these cations are particularly well represented in the Wadi Faynan area. Additional papers have indicated that lead can increase osteoporosis and it may disrupt the normal formation of calcium hydroxyapatite, thus critically weakening the bone (Skinner, 2000). Tandon et al. (2001), in their study, noted that effects of lead on humans include anaemia, abdominal colic, and gum wastage. These effects may have been common in ancient times in such a severely polluted landscape.

Thus, it can be seen that copper and lead, particularly when exhibiting synergism with other cations, can have profound effects on the health of humans, whether representative of ancient or modern populations.

3. Experimental procedure

Ancient human skeletal material was obtained; through official channels and permits, from burial sites adjacent to the ancient and derelict settlement of Phaino. These skeletons were in excess of 1500 years old for the Byzantine material and well in excess of 2000 years old for the Bronze Age material; they exhibit distinct evidence of arthritis, particularly of the limbs. The goat and sheep skeletal material was recent (ca. 10 years BP—before present); the older material (Table 2) was in poor condition and hence is entered as ''sheep/ goat'' rather than an attempt being made to determine the actual species involved. Control material of goats and sheep was collected from approximately 12 km away from the more obviously polluted mining and smelting spoil tip sites. In all cases, material of comparable size/age and condition was chosen. All skeletal material was dry at the time of collection; they were transported to the laboratory and maintained in individual triple-wrapped polythene bags. Samples were carefully maintained in a dust-free environment. In the laboratory the samples were sorted and dissected, where necessary, to obtain appropriate samples and to additionally remove superficial material. The samples were individually thoroughly washed with sterile deionized water and were subsequently dried, and then subsamples were prepared by acid digestion and then analyzed by means of a Perkin–Elmer 360 flame atomic absorption spectrophotometer (FAAS) in our laboratories at The Nottingham Trent University. A minimum of three replicates were employed in each case.

4. Results

The results of this investigation are presented in Tables 1 and 2. The samples from the Wadi Faynan area exhibit enhanced copper and lead values as compared with the material from the less polluted, more distant from the pollution sources, control site; nevertheless there exists evidence of clearly defined transport of these cations from the sources of pollution to the more distant

Table 1 Copper and lead content of modern samples (o10 years old)

Sample	Copper (mg/kg)	Lead (mg/kg)
Goat molar root	48	126
Goat molar root (control)	4	18
Goat molar crown enamel	20	109
Goat molar enamel (control)	4	49
Goat molar dentine	20	90
Goat molar dentine (control)	4	16
Goat jaw inner (articular) end	33	268
Goat jaw inner (articular) end (control)	12	20
Goat jaw distal (symphysial) end	28	270
Goat jaw distal (symphysial) end (control)	5	14
Sheep molar root	55	169
Sheep molar root (control)	8	26
Sheep molar crown enamel	22	129
Sheep molar enamel (control)	6	53
Sheep molar dentine	20	94
Sheep molar dentine (control)	6	46
Sheep jaw inner (articular) end	45	359
Sheep jaw inner (articular) (control)	17	43
Sheep jaw distal (symphysial) end	51	391
Sheep jaw distal (symphysial) end (control)	19	46
Asssociated sediment	31	16

Table 2 Copper and lead content of ancient samples Copper Lead (mg/kg) (mg/kg)

Sheep/goat molar root 61 326 Sheep/goat molar crown 49 271 Sheep/goat molar enamel 29 203 Sheep/goat jaw inner (articular) end 59 463 Sheep/goat distal (symphysial) end 45 524 Associated sediment 38 21 Bronze Age human femur (outer bone) 108 98 Bronze Age human femur (inner bone) 82 74 Associated sediment 37 21 Byzantine human femur (outer bone) 197 196 Byzantine human femur (inner bone) 177 170 Associated sediment 44 28 Byzantine human cranium (outer bone) 103 35 Byzantine human cranium (inner bone) 47 24 Byzantine cranium sediment contents 40 16 Ancient insect pupae from within thoracic cavity 30 11 Human rib adjacent to current position of 137 99 insect pupae sites (Table 1). Samples of teeth, lower jaws, and skeletons of goats, sheep, and ancient humans indicate that these organisms have bioaccumulated significant concentrations of both copper and lead from the environment; these patterns are explored more fully below. The molars of goats show a high concentration of copper (48 mg/kg) and lead

(126 mg/kg) in the root area, while lower values (Table 1) occur in the hard enamel (and the dentine) separated from the blood vessels, lymph vessels, and nerves occurring in the deeper pulp cavity. The values for copper in the lower jaw are lower than those obtained from the molar root area; however, in the case of lead, the values are markedly enhanced—e.g., goat jaw inner (articular) end possessed values of 268 mg/kg and the symphysial end 270 mg/kg. The values in the sheep teeth are also presented in Table 1. The molar root values for copper are slightly enhanced compared with those of goats and the lead value is also clearly enhanced. Again, these values are markedly enhanced as compared with the control values (Table 1). Comparison of portions of the molar with the lower jaw highlights the nature of both partitioning and bioaccumulation of these cations (Table 1).

Table 2 presents results from two graves, one Byzantine and one Bronze Age. The limited herbivore material associated with the Byzantine grave indicated marked bioaccumulation of both copper and lead in the various samples indicated. The values obtained indicated clearly defined bioaccumulation and partitioning and are examined in more detail below.

5. Discussion

Evidence is presented in Tables 1 and 2 that the samples of teeth and bone obtained from recent and older skeletons on the Wadi Faynan area of southern Jordan reflect the metalliferous mining history of the area in their enhanced copper and lead values. The values in the control material, collected from equivalent sites but lacking obvious pollution foci, are far less enhanced but indicate that pollutants, over several millennia, have dispersed out from the sites of mining and smelting to affect the more distant environment.

In the case of both the goat and sheep molars, the highest values for both copper and lead occur in the root area and not the enamel or dentine. This is a likely reflection that transport of such cations, followed by subsequent tissue deposition, is via the blood vessels of the pulp cavity and not penetration through the extremely hard and impermeable enamel. The values for copper are substantially lower than those for lead and it should be noted that copper is an essential element. The values for both copper and lead are generally lower in the goat than in the sheep samples examined. As these organisms occupy the same habitat, this is likely to reflect the fact that sheep graze and thence can be more readily exposed to heavy metals in the substrate, while goats also browse and hence may avoid some of the more heavily contaminated substrata. Thus, when teeth are used as indicators of environmental toxicology, in a comparative study, it is essential that comparisons involve identical portions of the teeth; analysis and chemical comparison of diverse portions is likely to generate erroneous results. This is an important consideration, as teeth are a valuable tool for the measurement of aspects such as environmental change and environmental pollution because they may persist in the ground for a prolonged period and hence, as is the case in this investigation, provide excellent vehicles for monitoring pollution in both recent and ancient times. The values of these cations are generally enhanced compared with their food sources; thus Pyatt et al. (2000) recorded values of lead and copper in the low-growing food plant Gymnarrhena micranthia of 40 and 60 mg/kg, respectively. Values obtained from the lower jaws of both sheep and goats, generally indicate much higher concentrations of copper and lead than the molars, which sit in sockets on the jaws. It is thus conceivable that molar teeth have many attributes that render them important toxicological indicators of ancient and modern environmental contamination.

The Bronze Age human femur samples showed enhanced values of both copper and lead compared with samples of the encompassing sediments. It is also apparent that partitioning of these cations occurs in such long bones, with values of both copper and lead being higher in the outer bone of the femurs. This, at first sight, is somewhat surprising considering the haemopoietic role of the bone marrow, but values may reflect possible routes of bioaccumulation including long-term deposition in the outer tissue of these long bones and/or contamination by heavy metals contained in the grave environment or during the course of life of the individual. Contamination whilst in the grave, post mortem diagenesis (essentially the exchange of ions between the bone and the soil), appears improbable as a consequence of the lack of real metabolic activity at this post mortem stage, together with the much lower values for copper and lead in the associated sediments as compared with femur values. Drought would also affect the solubility and mobilization of these pollutants. It is also interesting to note that the more recent Byzantine skeletal material (Table 2) contains values of both copper and lead much enhanced over the Bronze Age material. This is likely to reflect the fact that in the latter period, the extent of environmental pollution by copper and lead, as a consequence of preexisting metallurgical enterprises, would be enhanced. This is further supported by the values obtained for these cations in the grave sediments intimately associated with these skeletons. An additional point is that possibly, the individuals of the Byzantine period were more closely associated, on a longer term basis, with the mineral extraction activities. The values of copper in the samples examined were enhanced with the highest value at 197 mg/kg (Table 2); this represents a ca. 50-fold enhancement of copper as compared with the data of Scheinberg (1979), who produced a typical figure for copper in human bone of 4.2 mg/kg.

A portion of Byzantine human cranium was removed, prepared as formerly described, and analyzed. The copper and lead values (Table 2) were markedly enhanced compared with the cranium sediment contents; it may be noted that the external bone of the cranium contained enhanced values for copper (103 mg/ kg) and lead (35 mg/kg), with sediment values for copper and lead being 40 and 16 mg/kg, respectively. The values for the inner bone (adjacent to the former position of the brain) were significantly less, with copper at 47 mg/kg and lead at 24 mg/kg; i.e., copper and lead partitioning is evident in the human cranium. The higher values in the outer bone are unlikely, as noted earlier, to have developed post mortem but may reflect exposure of the skin, encompassing the cranium, during life to occupational metal pollution (e.g., metal-rich airborne particulates). Additional mechanisms, however, are conceivable. Baranowska et al. (1995) analyzed sternum bones (manubrium) obtained from individuals who had died in 1993 and had inhabited the grossly polluted Upper Silesian industrial region of Poland. Their values for copper and lead are generally lower than those reported above which reflects how polluted the Wadi Feynan area is and was in earlier times. Arnay-De-La-Rosa et al. (1998) measured lead concentrations in individuals, including those of the pre-Hispanic period and found much lower values (0.7–18.83 ppm) in these more recent skeletons. Arguably these data reflect the much lower values of environmental lead pollution on Tenerife as compared with southern Jordan, with its massive copper and lead exploitation record. Similarly, the values presented above are well in excess of those reported by Ahlgren et al. (1976) and Ahlgren and Mattsson (1979), who noted that the lead content of bones from metallurgical workers in Sweden was 40– 100 mg/g of bone.

Ancient insect pupae were recovered from the thoracic cavity of the Byzantine human skeleton and were found to have a copper content of 30 mg/kg and a lead content of 11 mg/kg. These values are far lower than those in associated rib bone (copper at 137 mg/kg and lead at 99 mg/kg, respectively). A number of points emerge from these findings and may be summarized as follows: (1) the ribs contain much lower copper and lead concentrations than the above-discussed femurs, (2) the values in the enclosed ancient pupae are much lower than those in the rib tissue, and (3) the values for copper and lead in the pupae are massively enhanced compared with values in modern invertebrates (Pyatt et al., 2002b). It may be suggested that the larvae, which eventually were to form the pupae, may have devoured contaminated human tissues and thus bioaccumulated cations.

6. Conclusions

Both the modern and ancient mammal samples exhibit massively enhanced copper and lead values derived from this copper-and lead-polluted area. The teeth potentially provide powerful tools to monitor long-term pollution, but only if consistency is ensured with regard to the material sampled. The values in the ancient human skeletons reflect the high pollution values in this area in the Byzantine Age and the Bronze Age; the partitioning in the cranium and in the femur is of particular interest. Care must hence be taken in sampling and analyzing such material to ensure that comparable material only is utilized.

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